Geochemistry of groundwater and saltwater intrusion in a coastal region of an island in Malacca Strait, Indonesia

Dewandra Bagus Eka Putra[1†], Mohamad Sapari Dwi Hadian[2], Boy Yoseph CSS Syah Alam[2], Yuniarti Yuskar[1], Wan Zuhairi Wan Yaacob[3], Bithin Datta[4], Widya Puspa Dwi Harnum[1]

[1]Department of Geological Engineering, Universitas Islam Riau, 28284 Pekanbaru, Riau, Indonesia
[2]Faculty of Geological Engineering, Universitas Padjadjaran, 45363, Jatinangor, Jawa Barat, Indonesia
[3]Geology Program, Universiti Kebangsaan Malaysia, 43600, Bangi, Selangor, Malaysia
[4]College of Science and Engineering, James Cook University, 4814, Townsville, Queensland, Australia

ABSTRACT

Most people in Bengkalis island use groundwater as their primary resource, but brackish water discovered in the wells located near the shore as well as in the distance. A hydrogeology study through geochemistry and isotope aspect conducted for the first time to identify the effect of saltwater intrusion (SWI) on groundwater. Na > Mg > K > Ca were the most dominant cation sequence in groundwater, while Cl > HCO_3 > SO_4 was the most dominant for anion. NaCl is the main type of groundwater in shallow aquifer and NaCl & $NaHCO_3$ both became the most dominant type of groundwater in the deeper aquifer system. Most of the groundwater samples from dug wells indicating the weak acid exceeding strong acid. Meanwhile, the groundwater samples from bore wells dominated by alkalis exceeding alkaline earth. SWI analysis shows 36.4% of groundwater samples from dug wells and 40% of bore water samples experiencing the salinization process. From isotope analysis, the groundwater samples found to be originated from the higher elevation or distance source. Meanwhile, there are lighter 2H values in the samples that are indicating the groundwater flown in the deeper part of the aquifer.

Keywords: Bengkalis, Coastal groundwater, Groundwater isotope, Hydrogeochemistry, Saltwater intrusion

1. Introduction

The people in developing countries often had dependency to groundwater resource and thus it becomes an essential and valuable water resource [1], the number of societies facing water shortages is increasing as the level of the groundwater table decreases as a consequence of excessive usage and the level of pollution of surface and groundwater resources increases due to several anthropogenic activities [2]. For this reason, there have been many studies on water quality in various parts of the world. As a vital source of drinking water and irrigation, groundwater quality should be monitor and protect from the contaminant [3]. Groundwater in the coastal area is commonly susceptible to an environmental problem such as saltwater intrusion [4, 5] and upconing [6]. The salinity of water resources is a significant problem that the world population suffers. It has been studied intensively during the past decades,

particularly in coastal aquifers [7]. The complexity and non-linearity of saltwater intrusion in the coastal area become a challenge [8], several reasons could explain the salinity of groundwater such as a consequence of leaching of evaporates in waterlogged areas, weathering of minerals, the movement of saline groundwater to the shallower aquifer as a result of over-pumping [9], change and variation of climate condition and rising of sea-level [10]. At this time, the coastal area facing rapid development. In several areas, the used of coastal land changed into the industrial area, which results in more problems not only for the groundwater but also seawater, especially in the area where the industrial waste has not managed improperly. Those could lead to groundwater quality problems, and even worse, the surrounding residents cannot use the resources in daily activity due to health issues [11].

Pumping test is a method to determine hydraulic properties [12], which could be used to identify the movement of the con-

taminant, the possibility of saltwater intrusion, and the aquifer capability as the primary source of water [13]. In addition, the electrical resistivity tomography (ERT) method could become a supporting data to model the geometry of aquifer [14] and locating contaminant [15]. This method could also detect the pathway of pollution [16] and the boundary of freshwater and saltwater from the difference of electrical current [17].

Groundwater conditions in coastal areas in Indonesia had been affected by the human, land used activity, and groundwork [18]. Seasonal differences affected the discharge volume and nutrient variations of groundwater [19]. The salinity of groundwater occurred in major cities and found to intruded the groundwater wells, both shallow and deep wells [20]. It is occurred not only on the wells that closed to the shoreline [21] but also in further sites [22]. One of the major provinces in Indonesia, Riau, has also faced saltwater intrusion and upconing problems, particularly in the coastal areas and in the islands. The study conducted in Bengkalis island (Fig. 1), located in the Malacca Strait on the east coast of Riau mainland. Initial groundwater monitoring had detected the salty water in freshwater wells as well as in the river [23]. The northern part of the study area occupied by people and there are several beaches for tourism spots, meanwhile in the land in the southern part of the study area occupied by the harbors, and populated residential area.

Stratigraphy condition of the study area (Fig. 1) based on the regional geological map from [24] was composed of Older Superficial Deposit (Qp) and Young Superficial Deposit (Qh). Older Superficial Deposits consist of clays, silts, clayey gravels, vegetation rafts, and Young Superficial Deposit consists of clays, silts, clean gravel, vegetation rafts, peat swamps [25].
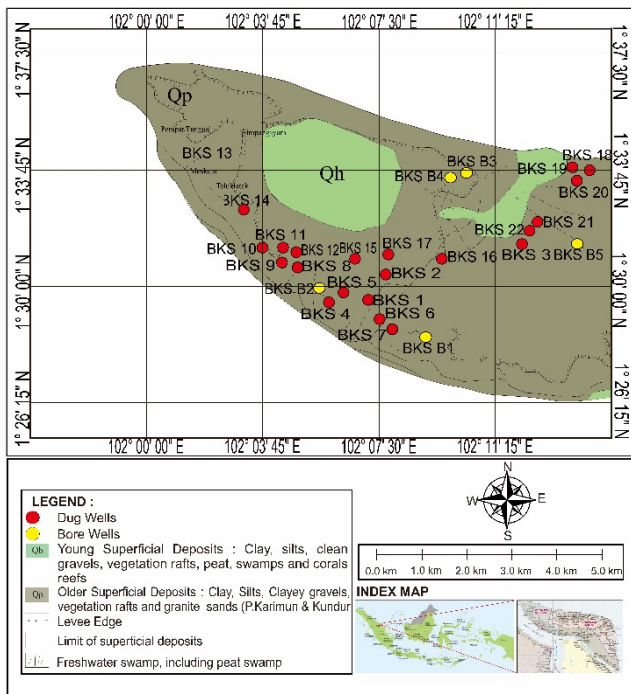

Fig. 1. Geological map of Bengkalis Island and well location map in the study area.

2. Methodology

Fieldwork activities consist of measuring the existing wells to obtain several data such as groundwater level, in-situ groundwater quality, and groundwater geochemistry (from water samples). Groundwater well was measured randomly both from shallow (dug) and deep wells across the island. In total, there are 22 dug wells and five bore wells (deep well) located near the coast and in the middle of the study area (Fig. 2). Five bore wells chosen by the availability of well's information (Table 1) as well as geological information of the well. The connection between dug well and bore well is the similarity of geological conditions surrounding the well.

Well's depth, electrical conductivity (EC), and total dissolved solids (TDS) were among the in-situ parameters that measured directly in the field. Water samples collected using a plastic bottle sealed with an inner cap, the bottle must be full with water so that no water bubbles inside, and preserved in the container to keep the water temperature in stable condition. The samples delivered to the Water Analytic Laboratory of National Nuclear Energy Agency (BATAN) to analyze the major ion such as Na, K, Ca, Mg, Cl, HCO_3, and SO_4. Stiff and piper diagram then plotted to determine the groundwater type.

Table. 1. Information on Groundwater Well in the Study Area

Well ID	Depth (m)	Average depth of water table (m)	Distance from the coast (m)
BKS 1	3.3	2.1	2,250
BKS 2	1.2	0.8	740
BKS 3	2.1	0.8	1,430
BKS 4	2.9	0.9	1,720
BKS 5	4.4	4.2	2,990
BKS 6	2.7	2.2	8,860
BKS 7	3.8	1	1,390
BKS 8	1.9	0.4	1,760
BKS 9	3.2	1.1	1,150
BKS 10	3	3	1,110
BKS 11	1.6	1.2	1,210
BKS 12	2.6	0.1	1,360
BKS 13	2.5	0.6	3,180
BKS 14	3	1.1	1,300
BKS 15	1.4	1.2	4,670
BKS 16	3.5	1.5	6,820
BKS 17	2.5	1.9	5,160
BKS 18	1.9	0.4	1,280
BKS 19	1.9	0.4	1,430
BKS 20	1.2	0.5	2,820
BKS 21	3	1	5,090
BKS 22	2	1	5,460
BKS B1	115	60	2,650
BKS B2	60	40	1,580
BKS B3	54	40	2,740
BKS B4	57	35	2,940
BKS B5	81	60	2,300

Saltwater intrusion analysis conducted using several methods commonly used in the previous study [26] to identified groundwater status:

a. The enrichment of Ca calculated using the formula from [27] and [28] based on the ratio of Ca and Mg concentration (in mg/L). High ratios (> 1) could be the indication of saltwater intrusion [27]

$$\text{Ca Enrichment} = \left(\frac{\text{Ca}}{\text{Mg}}\right) \qquad (1)$$

b. [29] proposed the Simpson Ratio based on the Cl, HCO_3, and CO_3 concentration (in mg/L).

$$\text{Simpson Ratio} = \frac{\text{Cl}}{HCO_3 + CO_3} \qquad (2)$$

The ratio describes the contamination level and divided into five classes as shown in Table 2.

Table 2. Groundwater Classes Based on Simpson Ratio

Ratio	Quality
< 0.5	Good quality
0.5-1.3	Slightly contaminated
1.3-2.8	Moderately contaminated
2.8-6.6	Injuriously contaminated
6.6-15.5	Highly contaminated

c. The ratio of sodium chloride (in mg/L) based on [27] to shows the impact on the groundwater. Saltwater intrusion impact would be shown by ratios less than 0.86, while anthropogenic sources of contamination would be demonstrated by ratio > 1. For the ratio value between 0.86 – 1, the mean value (0.93) become the limit to determine the contamination process. If it is less than 0.93, the contaminant source seems to be SWI. Meanwhile, if it is more than 0.93, the anthropogenic activity would be the cause of contamination.

$$\text{Sodium Chloride Ratio} = \frac{\text{Na}}{\text{Cl}} \qquad (3)$$

d. Calculating Base Exchange Indices (BEX) proposed by [30] to distinguish if an aquifer in the salinization or freshening process.

$$BEX = \text{Na} + \text{K} + \text{Mg} - 1.0716\text{Cl} \left(\text{in} \frac{\text{meq}}{\text{L}}\right) \qquad (4)$$

Positive BEX indicates the freshening process, negative BEX indicates the salinization process, and zero value indicates no base exchange.

Stable Isotopes (δ^2H, δ^2O) were analyzed using LGR Laser DLT-100. The result of stable isotopes plotted into the graph to identify the origin of the water. Interaction of groundwater-seawater traced using δ^2H and δ^2O isotope analysis and hydrochemical methods. Both isotopes used as a marker in identifying the source of groundwater in the aquifer, the possibility could be coming from rain, deeper or shallower groundwater, seawater, fossil water, or magmatic water ([21]; [31]). The isotope analysis in Riau province studied by [32] was the first to define the Local Meteoric Water Line (LMWL) found that the reference value for LMWL in this area is $\delta^2H = 7.6 \ \delta^{18}O + 10.5$ ($r^2 = 0.921$).

3. Result and Discussion

Hydrogeochemical analysis shows several major ions that have a significant role in the groundwater. The dominant ions found were sodium, chloride, bicarbonate, and sulfate. Carbonate analyzed, but the reagent shows no reaction indicated the absence of that ion. Table 3 shows the value for each major ion in groundwater samples.

The sequence of major ion could define the primary ions in a groundwater sample [33]. From groundwater samples that located near the coast, the cation sequences found as Na > Mg > K > Ca, Na > Ca > Mg > K, Na > Ca > K > Mg and Mg > Na > K > Ca, while Na > Ca > Mg > K, Na > Mg > Ca > K and Na > Mg > K > Ca sequences found in the samples located in the middle of the island. In general, the most dominant cation sequence is Na > Mg > K > Ca found in 11 samples.

The anion sequences found as Cl > HCO_3 > SO_4, Cl > SO_4 > HCO_3, HCO_3 > Cl > SO_4 dan SO_4 > Cl > HCO_3 for water samples that located close to the coast, while Cl > SO_4 > HCO_3 and HCO_3 > Cl > SO_4 sequences found in the samples located in the middle of island. Cl > HCO_3 > SO_4 is the most dominant anion sequence in the study area, found in 10 samples.

3.1. Groundwater Facies
The concentration of each major ion plotted into Stiff diagram (Fig. 2) and Piper diagram (Fig. 3) to identify the type and facies of groundwater in the study area.
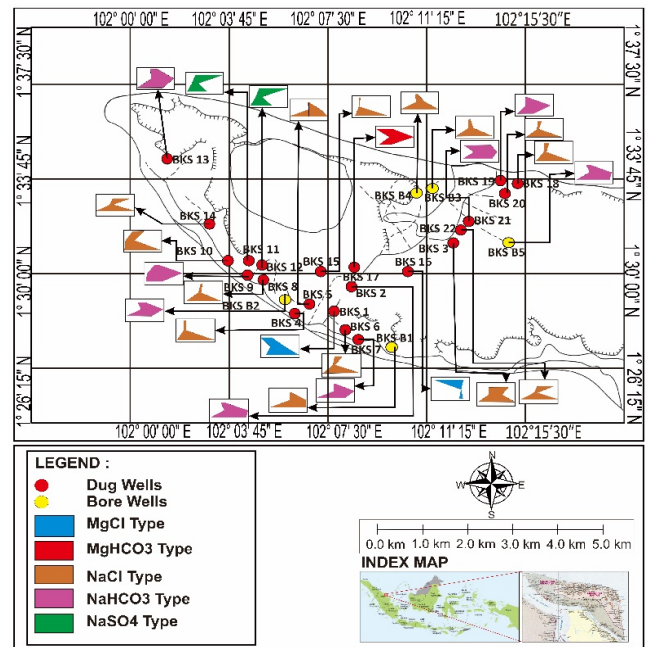

Fig. 2. Stiff diagram plot for groundwater in the study area shows the groundwater type in each location.

Table. 3. The Concentration of Each Major Ion in Groundwater Samples

Well ID	Parameters (Miligram/Liter)							
	Na	K	Ca	Mg	Cl	HCO$_3$	SO$_4$	CO$_3$
BKS 1	611.80	51.03	10.51	85.46	695.04	579.21	37.73	0.00
BKS 2	129.85	8.34	76.80	5.50	150.73	41.49	335.82	0.00
BKS 3	148.08	16.76	14.02	24.62	301.46	73.07	61.48	0.00
BKS 4	105.65	6.49	15.45	22.83	263.83	41.70	25.56	0.00
BKS 5	1,046.73	67.38	2.98	67.42	1,063.78	1,160.08	38.40	0.00
BKS 6	968.03	58.87	10.21	37.39	850.48	1,067.60	48.76	0.00
BKS 7	489.54	20.95	1.05	20.63	317.75	607.29	37.98	0.00
BKS B1	446.88	21.70	0.75	20.46	360.33	603.57	42.06	0.00
BKS B2	66.37	11.29	5.21	12.95	70.55	154.40	15.89	0.00
BKS 8	1,041.46	28.21	79.24	69.69	1,858.65	29.72	156.88	0.00
BKS 9	32.83	9.00	12.42	2.75	25.00	122.02	12.00	0.00
BKS 10	33.74	6.62	13.65	1.88	58.11	0.00	20.17	0.00
BKS 11	16.31	3.25	6.56	9.15	15.20	0.00	56.36	0.00
BKS 12	18.20	4.19	13.61	8.20	13.86	0.00	94.88	0.00
BKS 13	330.79	32.86	2.38	53.79	304.91	478.89	46.88	0.00
BKS 14	47.04	8.58	17.17	12.05	62.21	0.00	134.88	0.00
BKS 15	5.22	0.46	0.50	0.50	7.19	0.00	4.96	0.00
BKS 16	4.34	1.10	0.40	0.83	6.85	0.00	3.36	0.00
BKS 17	139.25	21.24	1.41	39.85	113.61	288.16	41.74	0.00
BKS 18	78.43	6.07	11.93	14.22	193.87	0.00	46.75	0.00
BKS 19	908.07	43.99	106.82	68.44	1,962.23	0.00	268.18	0.00
BKS 20	3,987.91	155.48	158.48	545.75	7,271.78	0.00	836.64	0.00
BKS 21	47.09	9.13	3.83	25.22	62.89	64.61	60.31	0.00
BKS 22	306.37	19.95	11.58	41.30	332.47	398.80	53.07	0.00
BKS B3	654.08	60.27	61.88	162.71	1,463.96	284.24	50.93	0.00
BKS B4	679.69	71.92	55.61	154.72	1,489.74	285.48	40.75	0.00
BKS B5	550.89	46.34	8.05	46.62	892.26	478.07	8.51	0.00
Maximum	3,987.91	155.48	158.48	545.75	7,271.78	1,160.08	836.64	0.00
Minimum	4.34	0.46	0.40	0.50	6.85	0.00	3.36	0.00
Average	477.58	29.31	26.02	57.59	748.47	250.31	95.59	0.00
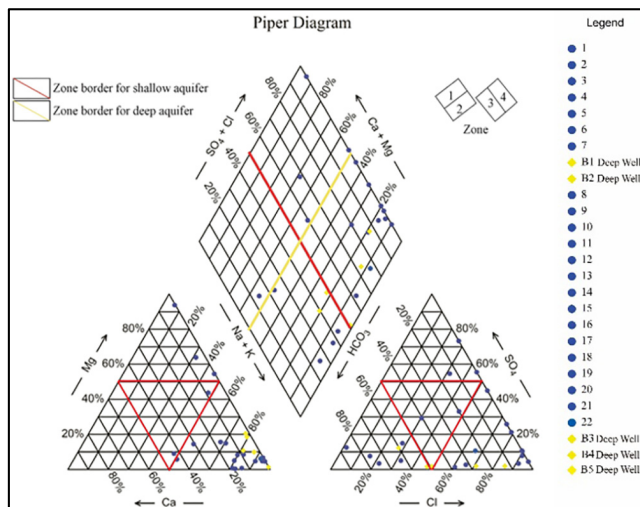
Fig. 3. Piper plot of shallow groundwater and deep groundwater samples.

Stiff diagram shows the water type based on dominant cation and anion in groundwater samples. Several water types had found, such as sodium chloride (NaCl), sodium bicarbonate (NaHCO$_3$), magnesium chloride (MgCl), magnesium sodium bicarbonate (Mg, Na-K, HCO$_3$) and sodium sulfate (NaSO$_4$). Sodium seems to be the governing cation to determined water type from groundwater samples, and for anion, chloride became dominant ions that are ruling in the determination of water types. Fig. 3 shows the distribution of dug wells groundwater samples and the distribution of deep wells groundwater samples using Piper diagram to identify the facies and evolution of groundwater in the study area.

There are several groundwater facies determined from the cation triangle (left side), such as magnesium type, sodium or potassium type, and no dominant type (shown by red triangle). Sodium or potassium type was the dominant type of groundwater from the cation plot. From the anion triangle (right side), there are sulfate type groundwater, chloride type, bicarbonate type, and no dominant

type (also shown by red triangle). Bicarbonate type is the most dominant type of groundwater from anion plot. The diamond diagram of Piper plot displays several groundwater facies such as calcium sulfate waters, sodium chloride waters, sodium bicarbonate waters, and calcium bicarbonate waters. The most dominant water facies is sodium bicarbonate waters found in 17 wells. In general, the shallow groundwater of the study area falls into zone 3 and zone 4. Zone 3 indicating weak acid exceeding strong acid and zone 4 indicating strong acid exceeding weak acid.

Groundwater samples in deep wells shown to be dominated by sodium or potassium type in the cation triangle. Meanwhile, both chloride type and bicarbonate type dominated in anion triangle. In combination, the piper diamond graph indicating the domination of both sodium chloride waters and sodium bicarbonates waters. Deep wells groundwater samples fall into zone 2, which points out alkalis exceeding alkaline earth.

3.2. Saltwater Intrusion Analysis

Several analyses had done to determine the status of groundwater toward the saltwater intrusion phenomenon (Table 4).

3.2.1. The enrichment of Ca
36.4% of shallow groundwater samples found to be affected by the saltwater intrusion having a ratio greater than 1. It comes from BKS 2, BKS 8, BKS 9, BKS 10, BKS 12, BKS 14, BKS 15, and BKS 19 station, while the rest of samples indicating normal condition by having a ratio less than 1. Meanwhile, the deep wells sample not yet reporting the saltwater intrusion problem.

3.2.2. Simpson ratio
From the calculation of chloride and bicarbonate ratio, 45.5% of shallow groundwater samples are fall into good condition category, 40.9% are in slightly contaminated, and 13.6% are in injuriously contaminated. Meanwhile, the deep groundwater samples found to be in good condition in BKS B2, slightly contaminated in BKS B1, moderately contaminated in BKS B5, and BKS B3, BKS B4 classified into injuriously contaminated state.

3.2.3. Sodium chloride ratio
Na/Cl ratio indicating the source of the contaminant in groundwater. 63.6% of shallow groundwater samples contaminated by saltwater intrusion (SWI), while the other 36.4% contaminated by anthropogenic sources. Deep well groundwater samples show saltwater

Table. 4. Saltwater Intrusion Analysis for Each Groundwater Well

WELL ID	Ca Enrichment	Remark	Simpson Ratio	Remark	Sodium Chloride Ratio	Remark	BEX	Remark	Overall Remark
BKS 1	0.122	Normal	1.199	Slightly	0.880	SWI	13.861	Freshening	More Freshening Process
BKS 2	13.963	SWI	3.632	Injuriously	0.861	SWI	1.740	Freshening	More SWI Process
BKS 3	0.569	Normal	4.125	Injuriously	0.491	SWI	-0.251	Salinization	More SWI Process
BKS 4	0.676	Normal	6.326	Injuriously	0.400	SWI	-1.365	Salinization	More SWI Process
BKS 5	0.044	Normal	0.916	Slightly	0.983	Anthropogenic	20.524	Freshening	More Freshening Process
BKS 6	0.273	Normal	0.796	Slightly	1.138	Anthropogenic	20.884	Freshening	More Freshening Process
BKS 7	0.050	Normal	0.523	Slightly	1.540	Anthropogenic	13.886	Freshening	More Freshening Process
BKS 8	1.137	SWI	62.538	Highly	0.560	SWI	-4.641	Salinization	More SWI Process
BKS 9	4.516	SWI	0.204	Good	1.313	Anthropogenic	1.126	Freshening	More Freshening Process
BKS 10	7.260	SWI	0.000	Good	0.580	SWI	0.028	Freshening	More SWI Process
BKS 11	0.716	Normal	0.000	Slightly	1.073	Anthropogenic	1.084	Freshening	More Freshening Process
BKS 12	1.659	SWI	0.000	Good	1.313	Anthropogenic	1.153	Freshening	More Freshening Process
BKS 13	0.044	Normal	0.636	Slightly	1.084	Anthropogenic	10.404	Freshening	More Freshening Process
BKS 14	1.424	SWI	0.000	Good	0.756	SWI	1.369	Freshening	More SWI Process
BKS 15	1.000	SWI	0.000	Good	0.726	SWI	0.061	Freshening	More SWI Process
BKS 16	0.481	Normal	0.000	Good	0.633	SWI	0.077	Freshening	More Freshening Process
BKS 17	0.035	Normal	0.394	Good	1.225	Anthropogenic	6.433	Freshening	More Freshening Process
BKS 18	0.838	Normal	0.000	Good	0.404	SWI	-1.145	Salinization	More Freshening Process
BKS 19	1.560	SWI	0.000	Slightly	0.462	SWI	-13.280	Salinization	More SWI Process
BKS 20	0.290	Normal	0.000	Slightly	0.548	SWI	1.700	Freshening	More Freshening Process
BKS 21	0.151	Normal	0.973	Slightly	0.748	SWI	2.449	Freshening	More Freshening Process
BKS 22	0.280	Normal	0.833	Good	0.921	SWI	7.147	Freshening	More Freshening Process
BKS B1	0.036	Normal	0.596	Slightly	1.240	Anthropogenic	10.744	Freshening	More Freshening Process
BKS B2	0.402	Normal	0.456	Good	0.940	Anthropogenic	2.100	Freshening	More Freshening Process
BKS B3	0.380	Normal	5.150	Injuriously	0.446	SWI	-1.038	Salinization	More SWI Process
BKS B4	0.359	Normal	5.218	Injuriously	0.456	SWI	-1.066	Salinization	More SWI Process
BKS B5	0.172	Normal	1.866	Moderately	0.617	SWI	1.910	Freshening	More Freshening Process

intrusion contamination in BKS B3, BKS B4 and BKS B5, and anthropogenic contamination in BKS B1, BKS B2.

3.2.4. Base exchange indices (BEX)

Salinization or freshening process in groundwater could identify using BEX value. The positive value indicating the freshening process shown by 77.3% shallow groundwater samples, and the other 22.7% shows the salinization process, which indicating from negative BEX value. BKS B1, BKS B2, BKS B5 shows freshening process in deep water well, while BKS B3 and BKS B4 indicated the salinization process.

3.2.5. Saltwater intrusion zone

The calculation methods show differences based on the result's similarity and the relationship between the techniques. Even though in several wells indicate the disagreement of the result compares
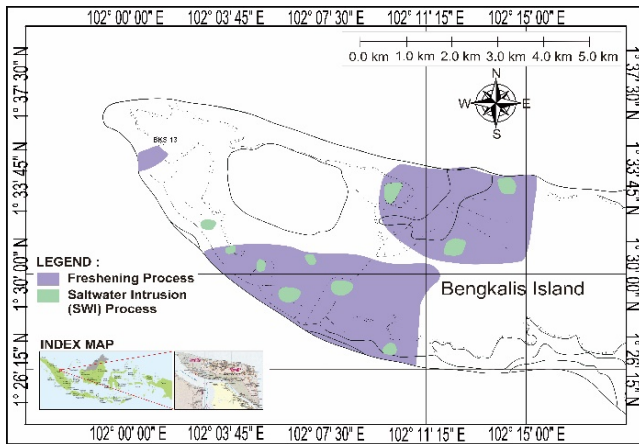

Fig. 4. The saltwater intrusion zone map in the study area shows the pattern of the saltwater intrusion zone.

to one method with another, in general, all these four methods are suitable to determine the saltwater intrusion problem in groundwater samples. Saltwater intrusion process found in 36.4% shallow groundwater samples and 40% of deep groundwater samples. The saltwater intrusion zone map (Fig. 4) shows the conditions of aquifer processing in the northern part and the southern part of the study area.

Most of the process was freshening of the aquifer and some indications of contamination from anthropogenic activity. However, several wells found to experience the saltwater intrusion process. The zone of intrusion likely to be upconing condition. The SWI impacted wells located in the middle of the freshening process, and the cause of this phenomenon could be excessive use of water.

3.3. Isotope Groundwater

Stable isotope δ^2H and δ^2O determined from 5 groundwater samples. They then plotted with Global Meteoric Water Line (GMWL) shown by a firm orange line and with Local Meteoric Water Line (LMWL) of Riau Province from [32] demonstrated by dash orange line.

The groundwater samples found to be in line with GMWL and LMWL, the water samples might be originated from the higher elevation or distance source. The more SWI process in the wells (Table 4) indicating the mixing of groundwater and seawater that intruded the wells, and it is an undergoing process. Meanwhile, BKS B4 and BKS B5 shown lighter 2H value compare to the other samples that are indicating the groundwater flown in the deeper part of the aquifer.

4. Conclusions

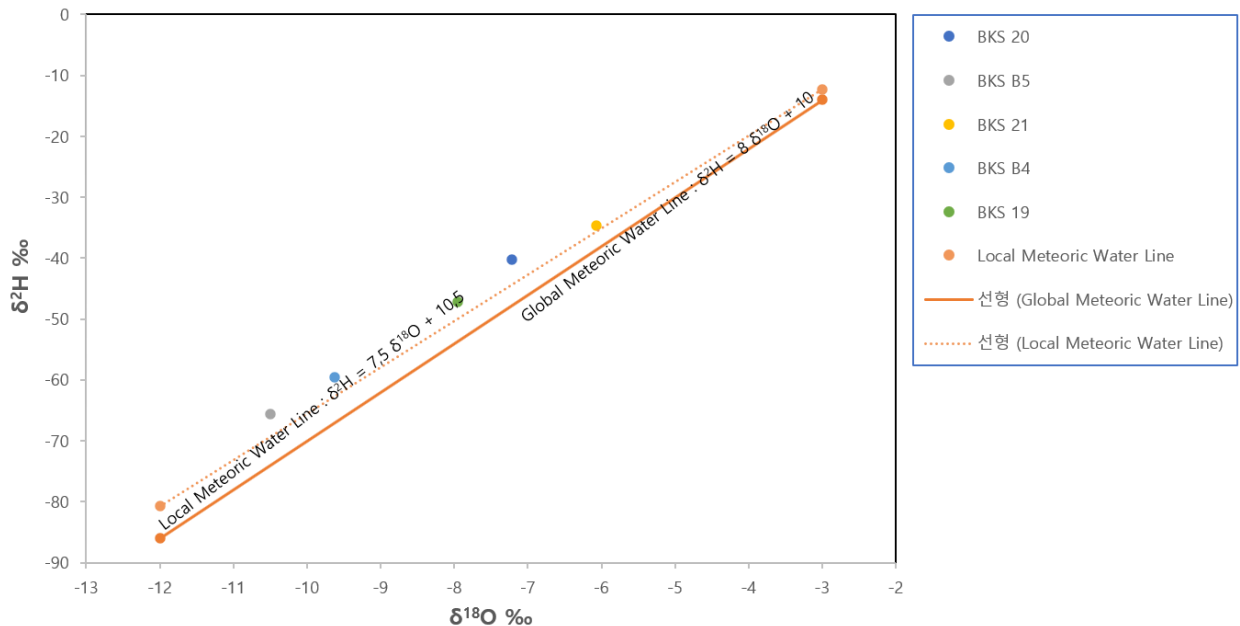In conclusion, the result of this research found several water

Fig. 5. Plot of groundwater isotope with global meteoric water line and local meteoric water line [32].

types in the study area determined from Stiff diagram, such as sodium chloride (NaCl), sodium bicarbonate (NaHCO$_3$), magnesium chloride (MgCl), magnesium sodium bicarbonate (Mg,Na-K,HCO$_3$) and sodium sulfate (NaSO$_4$). Also, Piper diagram shows the occurrence of several groundwater facies such as calcium sulfate waters, sodium chloride waters, sodium bicarbonate waters, and calcium bicarbonate waters. The identification of the saltwater intrusion problem using four different calculations shows a similar process in each groundwater well. Most of the groundwater aquifer experienced the freshening process. However, several wells suggested the saltwater intrusion impact and the mode of intrusion likely to be an upconing process. Isotope groundwater data in some wells give the information of water origin. The groundwater found originated either from the higher elevation or distance source, and there was an indication of deeper groundwater flow in the aquifer as well. The further study of isotope groundwater needs to perform to analyze groundwater-seawater interaction.

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Author Contributions

D.B.E.P (M.Sc) and Y.Y (M.Eng) conceived the study. D.B.E.P (M.Sc), M.S.D.H (Ph.D.), B.Y.C.S.S.S.A (Ph.D.) and W.Z.W.Y (Ph.D.) designed the research methods. D.B.E.P (M.Sc), W.P.D.H (B.Eng) and Y.Y (Ph.D.) data acquisition, M.S.D.H (Ph.D.), W.Z.W.Y (Ph.D.) and B.D (Ph.D.) provided statistical input; D.B.E.P (M.Sc), W.P.D.H (B.Eng) and B.Y.C.S.S.A (Ph.D.) data analysis, interpretation and wrote the manuscript. W.Z.W.Y (Ph.D.) and B.D (Ph.D.) provided grammatical revision to the manuscript. D.B.E.P (M.Sc) and W.P.D.H (B.Eng) provided manuscript revision.

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